Tellus (1994), 46B, 159–171
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TELLUS
ISSN 0280–6509

Modelling dry deposition of SO_2

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(Manuscript received 14 April 1993; in final form 23 November 1993)

ABSTRACT

The goals of this paper are to briefly describe experimental methods that are used to measure SO_2 dry deposition, and to discuss the physical, biological and chemical processes that control SO_2 deposition fluxes over vegetation and to describe how these fluxes are modelled. The predominant pathway for gaseous SO_2 uptake to dry vegetation is via turbulent transfer through the atmosphere surface boundary layer and molecular diffusion through the leaf laminar boundary layer and the stomata. The soil surface is a significant, but weaker sink for sulphur, especially when frozen or covered with snow. The appreciable solubility of SO_2 causes its uptake to be enhanced greatly in the presence of moisture on leaves and the soil. The aqueous uptake of SO_2, however, causes the pH of a solution to decrease which in turn produces a reduction in the solubility of SO_2. Neutralising species (ammonia, inner plant species) may cancel this reduction. A method is proposed to estimate local scale dry deposition fluxes of SO_2 in Europe. The method combines long-range transport modelling results, land use and surface specific data and an inferential approach.

1. Introduction

Sulphur dioxide was one of the first air pollutants recognised as harmful to humans and to ecosystems. Therefore, it is one of the most studied pollutants. Much research has been done in the early 70s. In the 80s, interest in Europe moved from sulphur compounds to nitrogen compounds. One of the main reasons for that, next to recognising nitrogen as being harmful, is that abatement strategies for sulphur emissions during the last decade have been successful in western Europe, Canada and the USA. Emission reductions in the order of 50% have been realised (Fig. 1). On regional scales both concentration and deposition of sulphur compounds decreased to the same extent.

Despite the large research effort put into the study of dry deposition of SO_2, no unequivocal process description is available. The factors influencing deposition are many and cause a very large natural variation in surface resistance's and deposition velocities. A problem arises when past research results are used for generalisation, because pollution climates under which measurements were made (surface wetness, humidity, ammonia) can often not be recovered. It is therefore a challenge to re-evaluate literature and combine the different results to obtain a clear and sound process description and parameterization.

It is not the purpose of this paper to present a summary of all the literature on SO_2 dry deposition, rather it is intended to summarise what is known and how to use this knowledge. Therefore, only a limited number of references have been included in the text. Much information presented here is based on some excellent reviews by Schwela (1977), Garland (1978), Chamberlain (1980), Voldner et al. (1986), Hicks et al. (1989), Davidson and Wu (1989), Wesely (1989) and Fowler et al. (1991).

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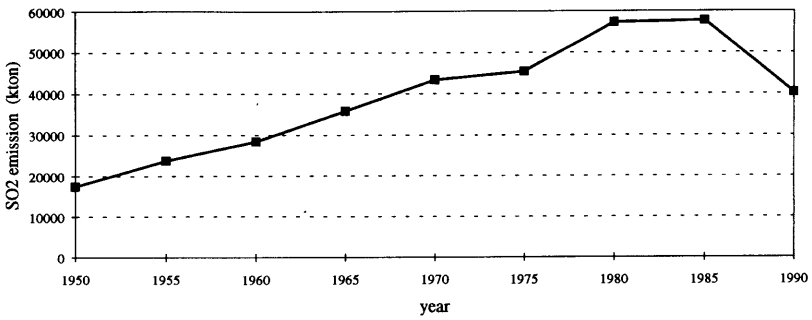


Fig. 1. Emissions of SO_2 in Europe 1950–1990 (kton a^{-1}). (Thomas et al., 1988; Iversen et al., 1991).

In this paper, a short overview of dry deposition processes will be presented. Then the state of knowledge will be discussed and gaps will be identified. Finally, recommendations will be given for dry deposition modelling.

2. Natural and anthropogenic sources of SO_2

Anthropogenic emission of SO_2 results from fossil fuel combustion. Most important sources are refineries, power plants, domestic heating and traffic. In addition to these anthropogenic sources, emissions of reduced sulphur compounds have also been quantified. Estimates of the amount emitted by the biosphere from oceans, soils, terrestrial vegetation as well as from vulcanos and biomass burning range between 35–70 Tg S a^{-1} (see Table 1). After oxidation, natural emissions are roughly equivalent to man-made emissions of about 70–100 Tg S a^{-1} on a global scale. In Northern America and Europe, however, anthropogenic emissions dominate.

Table 1. *Emissions of sulphur compounds to the atmosphere. Natural emissions as H_2S, COS, CH_3SH, CH_3SCH_3 DMS, and CS_2; anthropogenic sources as SO_2. All data in Tg (S) per year*

emissions from the oceans, mainly dimethylsulphide	16–38
soils and plants	4.8–15
vulkanism	9–13
coastal wetlands	2
biomass burning	>2.4
sum natural emissions	32–68
anthropogenic sources SO_2	70–100

Data from: Andreae (1990, 1991); Andreae and Andreae (1988); Andreae and Jaeschke (1992); Andreae and Raemdonck (1983); Bates et al. (1987); Berresheim et al. (1989); Cullis und Hirschler (1980).

3. Atmospheric chemistry

Once emitted, SO_2 may be oxidised to sulphuric acid aerosols or sulphates by reactions occurring in the gas phase, in the liquid phase, on the surfaces of solids, or combinations of these three. Rates of conversion generally seem to be larger during the day than at night, and in summer compared to winter. Additionally, the presence of liquid water in aerosols, clouds or fog is accepted to be an important factor in determining the rate of conversion of SO_2 (Finlayson-Pitts and Pitts, 1986). Furthermore, the reaction rate in liquids is influenced by the pH, metallic catalysts and oxidising pollutants (O_2, O_3, H_2O_2).

Thus sunlight intensities, presence of oxidants and/or oxidant precursors, humidity and the presence of fogs and clouds all appear to affect conversion rates. Because of the influence of these variable factors, a wide range of reaction rates is reported in the literature (see Finlayson-Pitts and Pitts, 1986 for a review). Typical summertime oxidation rates in rural areas cause 0.1 to 1.5% loss of SO_2 per hour.

4. Dry deposition of SO_2

In the atmosphere, sulphur compounds are ultimately lost through dry, wet and fog/cloud deposition. Mechanisms of dry deposition of SO_2

have been examined in laboratory experiments and in field studies. These studies show that once transported by turbulence and molecular diffusion to vegetation, SO_2 is absorbed readily by stomata. Uptake of SO_2 by external leaf surfaces and the soil can be a major sink in the presence of moisture. Most field studies report deposition velocities (V_d), i.e., the ratio between the flux F and the ambient concentration $C(z)$:

$$V_d(z) = \frac{F}{C(z)}. \quad (1)$$

Because $C(z)$ is a function of height z above the ground, V_d is also a function of z, and should be related to a reference height. F is assumed to be constant over the appropriate range of heights. The use of the concept of V_d is valid for SO_2 because its flux is nearly always uni-directional (downwards) and the surface is a sink for SO_2. The goal of many early studies was to support modelling efforts by quantifying the magnitude and variability of V_d over a variety of surfaces.

4.1. Dry deposition measurements

SO_2 dry deposition measurements have been made using several methods (for a discussion on measuring methods, see Erisman et al. (1993b)). These can be separated into micro meteorological methods, surface analysis methods and chamber methods. Micro meteorological methods comprise the eddy correlation method, the gradient method, variance methods, conditional sampling and eddy accumulation. The first two are most used. These methods are mainly used to quantify the short-term flux and to determine the regulating processes (meteorology, type of surface and surface conditions, and vegetation behaviour). There are several important limitations to these methods (Businger, 1986; Baldocchi et al., 1988; Fowler and Duyzer, 1991).

Surface analysis methods include analysis of snow, leaf washing techniques and the throughfall technique. The throughfall technique measures the total sulphate flux beneath the canopy. Several assumptions have to be made to estimate SO_2 dry deposition from these measurements. This method is well suited for deposition measurements where micro meteorological methods cannot be applied (complex terrain, forest edges). Comparison between results of throughfall measurements and other methods is presented in, e.g., Erisman et al. (1993b).

Chamber methods in which gas uptake by the depositing surface is measured are used in the laboratory or in the field. In open-top chambers or in closed chambers the characteristics of the vegetation is studied by controlling the environment. Factors thought to affect the plant are carefully controlled and measured. The response of different species of plants or different plant components can be studied in semi-isolation. The deposition to the surfaces (soil or vegetation) in the chambers can be estimated by comparing the in- and out-flux of the chamber over a certain time interval (e.g., Milne et al., 1979; Taylor et al., 1983; Granat and Johansson, 1983). In addition, cuvette and mini-cuvette studies with branches or single leaves, provides information into fundamental physical, chemical and biological processes related to trace gas exchange. There are several important limitations to these methods (Murphy and Sigmon, 1989).

4.2. Deposition velocity and resistance to transport

In the literature, V_d ranges are reported of 0.1 to 2 cm s^{-1} (conifer forests[1]; deciduous forests[2]; heath land[3]; crops[4]; grasslands[5]; water bodies[6] and bare soil[7]). Daytime values over a variety of plant surfaces are on the order of 0.8 to 1.2 cm s^{-1}[8]. Greater values (exceeding 2 cm s^{-1}) are observed over water surfaces[9] and forests[10] and

[1] Galbally (1979); Garland (1977); Hicks *et al.* (1982); Fowler and Cape (1983); Lorentz and Murphy Jr. (1985); McMillen *et al.* (1987); Erisman *et al.*, (1993d).

[2] Petit *et al.* (1976); Matt *et al.* (1987); Meyers and Baldocchi (1987).

[3] Erisman *et al.* (1993c).

[4] Fowler (1978); Fowler and Unsworth (1979); Hicks *et al.* (1989).

[5] Garland (1977); Platt (1978); Van Dop *et al.* (1983); Hicks *et al.* (1983; 1986); Neumann and den Hartog (1985); Duyzer and Bosveld (1988); Davies and Mitchell (1983); Davies and Wright (1985); Erisman *et al.* (1992).

[6] Welphdale and Shaw (1974); Garland (1977).

[7] Garland (1977).

[8] Garland (1977); Weseley and Hicks (1977); Nicholson and Davies (1988); Erisman *et al.* (1992).

[9] Whelphdale and Shaw (1974).

[10] Vermetten *et al.* (1992); Erisman *et al.* (1993d).

smaller (<0.13 cm s^{-1}) values are measured over snow[11] and bare soil[12].

V_d is dependent on many variable parameters, such as wind speed, temperature, radiation, etc. and surface conditions. The transport of pollutants from the atmosphere to the ground comprises three stages: first, the materials must be transported through the atmosphere to the receptor surface (turbulent layer); second, there is transport through the quasi-laminar layer at the surface; last, the gas (or particle) must be captured by the surface. To describe the surface exchange processes a common framework is provided. In this framework, the resistance analogy (Fig. 2), particularly the three stages in dry deposition to the ground are separated (Thom, 1975; Garland, 1977; Wesely and Hicks, 1977; Fowler, 1978): the aerodynamic resistance R_a, the quasi-laminar resistance R_b, and the canopy or surface resistance R_c. The inverse sum of the resistance's gives the deposition velocity:

$$V_d(z) = \frac{1}{R_a(z) + R_b + R_c}. \quad (2)$$

The aerodynamic resistance determines the rate of transport of gases between a given level in the atmosphere and the height of the effective surface sink. R_a can be estimated from measurements of wind speed, temperature and radiation and on knowledge on the roughness length. R_b is associated with the transfer through the quasi-laminar layer in contact with the surface; it quantifies the way in which pollutant transfer differs from momentum transfer in the immediate vicinity of the surface. R_b can be estimated from the friction velocity u^* and from component specific constants. R_c values presented in the literature are usually based on measurements of V_d, and is commonly determined as a residual resistance. By determining R_a and R_b from the meteorological parameters, R_c can be calculated as the residual resistance using eq. (2). R_c has been shown to be very variable. Its value is often quite uncertain since errors in the measurements accumulate in this term. However, in ideal conditions values of R_c can be related to surface conditions, time of day, season, etc., yielding parameterizations (Subsection 4.3).

The resistance analogy has been formalised in a 1-D model for estimating dry deposition rates (Hicks et al., 1987). In this "Big-leaf" model, the most important processes that are known to control dry deposition are parameterised. The flux is estimated from the concentration, wind speed, temperature, radiation, roughness length and R_c. R_c values can be estimated from theoretical considerations based on, e.g., solubility and equilibrium calculations in combination with simulations of vegetation specific processes, such as accumulation, transfer processes through stomata, mesophyll and cuticles, absorption, etc. R_c values thus obtained are, however, hard to validate by measurements because of the complexity of the processes involved.

4.3. Surface resistance

Numerous field studies show that the surface resistance R_c is the major controller of SO_2 deposition. R_c is the most difficult to describe of the three resistance's. R_c is a function of the canopy stomatal resistance R_{stom}, the canopy cuticle resistance $R_{cuticle}$, and the soil resistance R_{soil}. In turn, these resistance's are affected by leaf area, stomatal physiology, soil pH, and the presence and chemistry of liquid drops and films. The stomatal, leaf surface (cuticle) and soil resistance's act in parallel (see Fig. 2):

$$R_c = \frac{1}{R_{stom}} + \frac{1}{R_{soil}} + \frac{1}{R_{cuticle}}. \quad (3)$$

Physiological control. Most SO_2 enter plants through the stomata. As gas molecules enter the leaf, deposition occurs as molecules react with the moist cells in the sub-stomatal chamber and the mesophyll. Several conclusions can be drawn about environmental effects and inter-specific differences on SO_2 deposition based on known stomatal physiology. Thus, factors that increase stomatal resistance should increase the SO_2 R_{stom}. Stomatal resistance decreases hyperbolically with increasing light and increases linearly with increasing vapour pressure deficits (Jarvis, 1976). Soil water deficits cause stomata to close after some threshold deficit level is exceeded. Low and high temperatures cause stomatal closure and moderate temperatures promotes stomatal opening. Leaf

[11] Barrie and Warmsley (1978), Welphdale and Shaw (1974), Cadle *et al.* (1985), Granat and Johansson (1983), Valdez *et al.* (1987); Davidson and Wu (1990).

[12] Payrissat and Beilke (1974); Garland (1977).

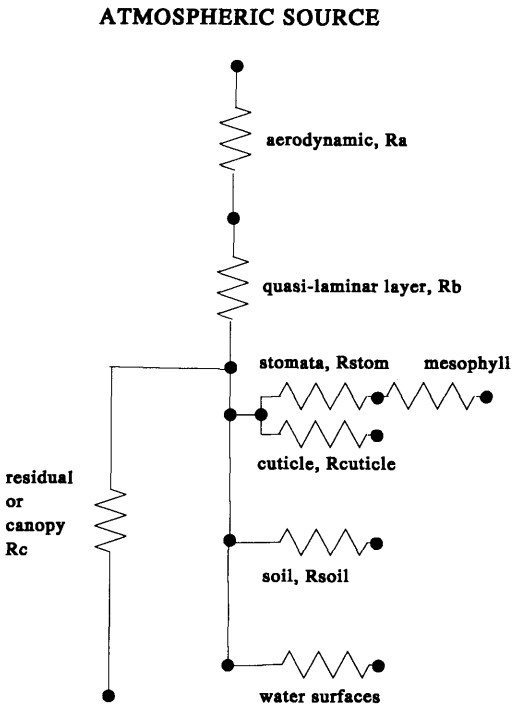


Fig. 2. Resistance diagram for the transport of SO_2 to various types of surfaces, taken from Hicks et al. (1987).

age, nutrition and adaptation are other factors affecting stomatal resistance. Elevated exposure to SO_2 causes stomata to close (Black, 1982). Stomatal resistance is different for different types of vegetation. Values of R_{stom} under ideal conditions during daytime, range between 30 and 300 s m^{-1} for a range of herbaceous annuals and woody perennials (Fowler, 1985; Baldocchi et al., 1987; Matt et al., 1987).

The canopy cuticle resistance far exceeds the canopy stomatal resistance; $R_{cuticle}$ ranges between 3000 and 40000 s m^{-1} (van Hove et al., 1989). However, it is observed that the surface resistance decreases as relative humidity increases (van Hove et al., 1989; Garsed, 1985; Erisman et al., 1993a; Erisman and Wyers, 1993; Draaijers and Erisman, 1993). This might indicate that the cuticle resistance is lowered at increasing humidity (van Hove et al., 1989) or that the surface becomes moist at increasing humidity, i.e., through deliquesce of salts at the external leave surface. When the cuticle is wetted or affected by pre-deposited particles or gases, SO_2 deposition to the surface also occurs via absorption and via chemical oxidation reactions with the surface.

Physical-chemical control. SO_2 dry deposition is enhanced over wet surfaces (Garland and Branson, 1977; Fowler and Unsworth, 1979; Fowler, 1985; Vermetten et al., 1992; Erisman et al., 1993a; Erisman and Wyers, 1993); the absorption is associated with a series of reactions (Seinfeld, 1986):

$$\begin{aligned} &SO_2(g) + H_2O \Leftrightarrow SO_2 \cdot H_2O \\ &SO_2 \cdot H_2O \Leftrightarrow H^+ + HSO_3^- \\ &HSO_3^- \Leftrightarrow H^+ + SO_3^{2-}. \end{aligned} \quad (4)$$

The total dissolved $S(IV)$ in chemically clean solutions is related to the partial pressure of SO_2 over the solution (p_{SO_2}) and the pH of the solution:

$$[S(IV)] = K_H \cdot p_{SO_2} \left(1 + \frac{K_1}{[H^+]} + \frac{K_1 \cdot K_2}{[H^+]^2}\right). \quad (5)$$

K_H is the Henry coefficient for SO_2 (5.4 mol l^{-1} atm^{-1} at 15°C) and K_1 and K_2 are the rate coefficients for reactions 1 and 2, respectively. From eq. (5) it can be concluded that an increase in acidity will reduce the amount of dissolved SO_2. The oxidation of $S(IV)$ to $S(VI)$ by O_2, O_3 and H_2O_2 provides another source of protons. The oxidation of aqueous SO_2 by O_3 and H_2O_2 is fast, oxidation by O_2 is slow unless there is a metal catalyst (iron or manganese). An increase in acidity due to solution and oxidation of SO_2 in water films will reduce SO_2 dry deposition to wetted surfaces. The presence of acid neutralising species either from the atmosphere (base cations, NH_3) or from the inner parts of the leaves, will reduce the acidity of the water film, abolishing the SO_2 dry deposition reduction. At present too little information is available to quantify these processes in terms of the influence on the magnitude of the surface resistance.

As the pH of the wetted surface is the deposition rate limiting factor, the origin of surface wetness is important: by fog, cloud, rain or dew, guttation (Fowler and Unsworth, 1979). The pH of rain, cloud and fog is less than that of pure water. The chemistry of wetted surfaces is affected by uptake of acidic precursors and oxidants, particles on the leaf surface, chemical lifetime, droplet size, leaching of nutrients from leaf and evaporation

rate of wetness (Brimblecombe, 1978; Chameides, 1987; Wesely et al., 1990).

The deposition of NH_3 neutralises an acid solution. Thus simultaneous deposition of SO_2 and NH_3 should experience higher SO_2 (and NH_3) dry deposition rates to water surfaces than would be found by SO_2 (or NH_3) alone (Wesely et al., 1990; Erisman and Wyers, 1993; Erisman et al., 1993c). Van Hove et al. (1989), Fowler et al. (1991) and Erisman and Wyers (1993) present evidence, showing that the surface SO_2 uptake at wet surfaces is enhanced in the presence of NH_3. This process is generally referred to as co-deposition. One site of reaction is in the water-filled pores of the cuticle at relative humidities above about 70% and the other site is on the wetted leaf surface.

Deposition to soil and litter. Deposition to canopies involves deposition to both vegetation and soil. Early studies assumed that deposition to soils under vegetation was relatively small (5–10% of the total flux; Fowler, 1979). Recent work shows that a substantial amount of material can be deposited to the soil below vegetation. Meyers and Baldocchi (1992) report 20 to 30% of SO_2 depositing on a deciduous forest is received at the forest floor. This substantial transfer occurs because large-scale intermittent eddies are able to penetrate through the vegetation and transport material to the soil. Differentiation of deposition to the canopy and to the soil is difficult to measure. Furthermore, it may be variable both in time and for different canopies.

Deposition to soil decreases at soil pH below 4 and increases with relative humidity (Garland, 1977; Payrissat and Beilke, 1974). When surface temperatures fall below zero or the surface is covered with snow, R_c values increase up to values of 200–500 s m^{-1} (Voldner et al., 1986; Erisman and Wyers, 1993). The deposition of SO_2 to snow covered surfaces depends on pH, snow temperature and probably the amount of SO_2 already scavenged by the snow pack (Caddle et al., 1985; Hicks et al., 1989).

5. Modelling regional scale SO_2 dry deposition

Currently two types of model are used to assess regional scale SO_2 dry deposition; i.e., long-range transport modelling and inferential modelling. In the long-range transport models deposition is studied from the atmospheric point of view, in order to find out how much is still left in the atmosphere to be dispersed further. In inferential models deposition is studied from the receptor point of view, to determine the flow of pollutants into the various ecosystems. For both purposes, reliable deposition monitoring or estimation methods are necessary. However, accuracy and spatial resolution needs may differ. The dry deposition parameterizations in the long-range transport models are not very detailed, whereas the inferential type of modelling contains a more detailed dry deposition process description.

Dry deposition velocities of SO_2 in long-range transport models are obtained from field and laboratory measurements. They are often independent of receptor surface. A meteorological correction is applied by introducing daily and seasonal variation (Iversen et al., 1991), or by describing V_d in terms of resistances (van Jaarsveld and Onderdelinden, 1992). Such parameterizations fail to make full use of the available understanding of dry deposition processes.

In inferential or "Big-leaf" models, dry deposition to relatively simple surfaces is inferred from the concentration of the chemical of interest and the appropriate V_d (Hicks et al., 1987). The concentration can be estimated from measurements or from long-range transport models. Surface characteristics and conditions are incorporated into the multiple-resistance transfer model used to calculate V_d (eqs. (1) and (2)). The state of knowledge on the most important parameters in the dry deposition process determines the details in the resistance description.

Resistance algorithms for gaseous uptake by individual leaves have been used by O'Dell et al. (1973), Murphy et al. (1977), Baldocchi et al. (1987, 1988) and Meyers (1987) to extend computations of SO_2 deposition to leaves to the canopy scale. The leaf surface resistance contains the parallel resistances exerted by the stomata and cuticle. Stomatal resistance can be computed using a multiplicative phenomenological model (Jarvis, 1976; Baldocchi et al., 1987) or a photosynthesis-driven model (Collatz et al., 1991). The internal concentration and the mesophyll resistance for SO_2 transfer are often zero (O'Dell et al., 1973; Taylor and Tingley, 1983). Empirical relationships

are usually used to specify cuticle resistance. The most pressing current limitation to our understanding of SO_2 uptake by vegetation is leaf surface uptake, its variability and the chemical processes taking place on the surfaces.

These models have some important drawbacks, e.g., that the model framework is designed only for deposition. For those components which can be deposited to and emitted from the surface, this framework does not apply.

6. Identification of major uncertainty sources in current regional scale estimates

The uncertainty in regional scale deposition estimates strongly depends on the pollution climate and on landscape complexity of the area under study. The uncertainty is determined by the major source of input, i.e., wet, dry or cloud and fog deposition. Furthermore, deposition estimates yield higher uncertainty in areas build up by complex terrain and in areas with strong horizontal concentration gradients. In Lövblad and Erisman (1992) a table has been presented to identify the uncertainties associated with major key factors in deposition for seven different (pollution) regions in Europe. In this table only nitrogen compounds were given; in Table 2, this is done for SO_2.

This is only a crude estimate of uncertainty; because of the strong local variation of dry deposition, local scales should be considered.

7. Synthesis and recommendations

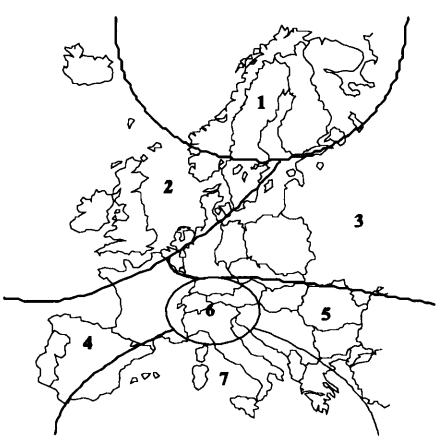
Up to now, transport models have been used to estimate long-range transport of air pollution and to produce so-called blame matrices in Europe. On the other hand, small scale models have been developed for process description and explanatory purposes. Recently, a third objective has become important, i.e., the modelling of local scale (ecosystem level) deposition fluxes. In contrast to long-range transport models, local scale models better include the large spatial variability of (dry) deposition. Local scale deposition fluxes are used for correlative research on deposition and possible effects, and for estimating exceedances of critical loads at the appropriate level. Some models have been developed for this purpose on a national scale

Table 2. *Uncertainty of key factors influencing deposition estimates of SO_2 in different pollution regions in Europe; the distribution of the numbered regions is given in the small map of Europe*

Regions:	1	2	3	4	5	6	7
Key factors							
emission/type	−	+	+ +	+	+	+	−
of source	1	1	3	1	3	1	1
concentration	+ +	+ +	+ +	+ +	+ +	+ +	+ +
	1	2	3	3	3	2	2
wind speed	+	+ +	+ +	+	+ +	+ +	−
	1	1	1	1	1	1	1
roughness length	+	+	+	+	+	+	+
	1	1	2	2	2	2	2
surface wetness	+	+ +	+ +	+	+ +	+ +	+
	3	2	3	2	3	3	3
orography	+	−	+	+	+	+ +	−
	2	1	2	2	2	2	1
co-deposition	−	+ +	+ +	+	+	+	+
	3	2	3	3	3	3	3
surface resistance	+ +	+ +	+ +	+	+ +	+	+
	3	3	3	3	3	3	3
dry deposition	−	+ +	+ +	+	+ +	+	−
	3	3	3	3	3	3	3
wet deposition	+ +	+	+	+	+	+ +	+
	1	1	1	1	2	2	2
cloud and fog	+	+ +	−	−	+	+ +	−
	3	3	3	3	3	3	3

legend: importance of key factors — uncertainty

+ +	dominating	1 low	<30%
+	important	2 median	30–70%
−	neutral	3 high	>70%
—	unimportant		



(UK Review Group on Acid Rain, 1987; 1991; Erisman et al., 1989; Erisman, 1992, 1993; see also Lövblad and Erisman, 1992). For Europe as a whole, no model is available yet.

Even though it is desirable to estimate dry deposition in Europe merely on measurements, this is not practical and would be prohibitively expensive. It is recommended to use a combination of models and measurements to map current dry deposition. Measurements are used to derive process descriptions and component and surface specific parameters. Furthermore, measurements are needed for evaluation of models and for establishing trends (Erisman et al., 1993b). Based on the current state of knowledge, SO_2 dry deposition on a local scale in Europe may be estimated by combining results of long-range transport models with a deposition module based on the inferential technique (Erisman, 1993; Van Pul et al., 1992). The principle of the proposed method to be used is visualized in Fig. 3.

7.1. Regional scale

With present state of knowledge, it should be possible for long-range transport models to map SO_2 concentration and deposition over Europe on

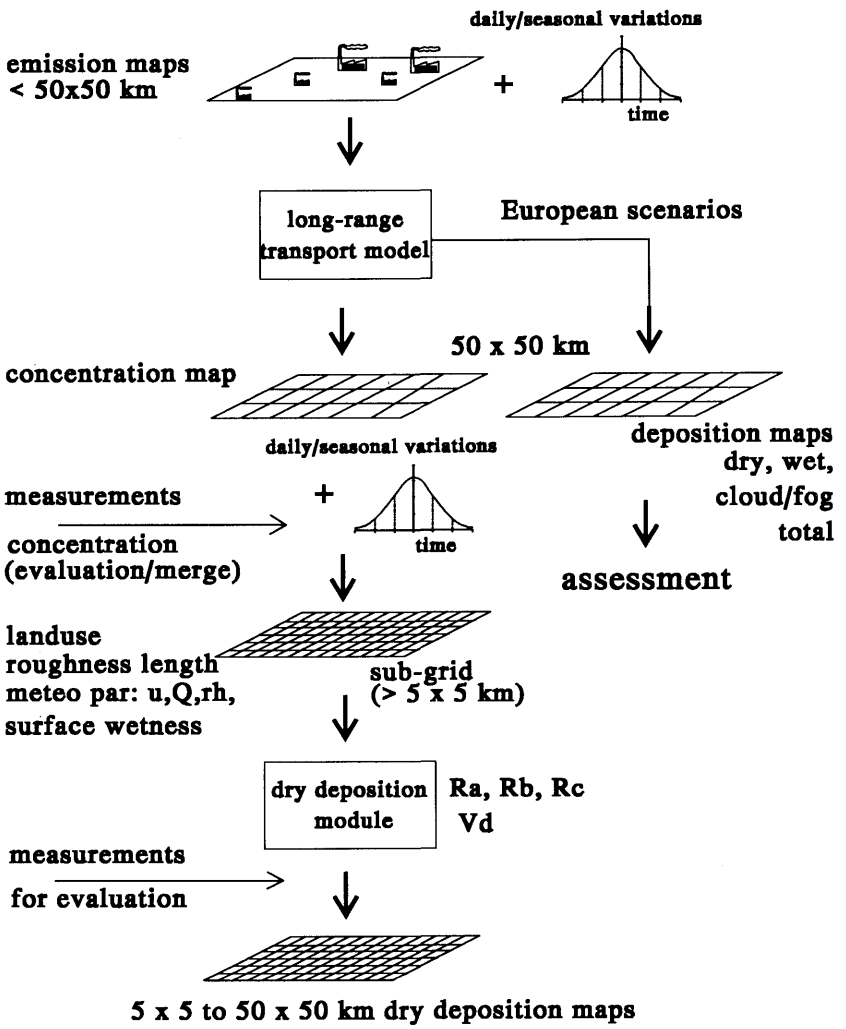


Fig. 3. Calculation scheme for dry deposition of SO_2 to simple surfaces.

a 50×50 km scale, provided that emissions are available on at least a 50×50 km resolution. The long-range transport model results can be used directly for assessment studies, scenario studies and concentration and deposition mapping over Europe. The dry deposition parameterizations in these models have to be roughness length and R_c dependent. Roughness length and R_c maps used in the model must be composed of subgrid maps (in time and space) used in the inferential dry deposition module.

7.2. Local scale

Local scale or sub-grid dry deposition maps can be made by using the long-range transport concentration and wind speed maps as input for a dry deposition module based on the inferential technique. The concentration and wind speed maps should be representative for a height between 50 and 100 m above the soil. At this height, still within the surface (constant flux) layer, concentration and wind is more or less independent of the underlying surface roughness. The concentration and wind speed maps should be of annual average values with parameters describing the seasonal and daily variation for each grid. Temporal variations might be described from model results itself or can be obtained from measurements (EMEP monitoring network; national networks). The concentration maps might be tested with measurements or merged with measurements. The wind speed maps can be obtained from synoptic measurements (the WMO synopsis database achieved by the European Centre for Meteorology and Weather Forecasting, ECMWF) or from whether forecast models, covering the whole of Europe.

Other input data for the deposition module are land use and roughness length maps, data on temperature, relative humidity and global or net radiation, and data from which surface conditions can be obtained, such as snow cover, frost, wetness, interacting species, water stress, soil pH, etc. In the deposition module the dry deposition parameters are coupled to the land use datasets.

The deposition module is based on a description of the dry deposition process in terms of the resistance analogy (Fowler, 1978; Hicks et al., 1987; Erisman et al., 1993c, see Section 4). The final output of the module is the SO_2 dry deposition flux per land use type on a minimum grid size of 5×5 km. This is the smallest grid, because for smaller grids, the roughness length concept is not applicable. The roughness length maps are used, together with wind speed, temperature and radiation data, for estimation of the friction velocity u^*, R_a and R_b. The land use, temperature, relative humidity and radiation data, and data on surface conditions are used to estimate R_c values. The parameterization of R_c should be based on available knowledge as discussed above.

The time steps used in the deposition module depend on the dry deposition process itself. Strong variations in dry deposition occur during the day and during seasons, because of stomatal behaviour and variations in concentrations (emissions). Furthermore, strong variations are due to the influence of surface wetness. These variations require hourly estimation of the dry deposition flux. This might be done for certain countries in Europe, where hourly data are available. However, for the whole of Europe, 6 hourly average data are available of most parameters from the ECMWF. It is expected that these data will serve to include daily variations.

The fluxes obtained from the deposition module should compare within certain limits with the long-range transport model results on an annual base for 50×50 km grids; i.e., on these scales z_0 and R_c values are comparable. This might be a first check on the deposition module (local scale) results. Furthermore, a check on mass consistency of these results is obtained. Evaluation of results obtained using this method with measurements can be done in several stages and on different scales (Erisman et al., 1993b). Next to evaluation of model results, an extensive uncertainty analysis should be made. The uncertainty analysis will serve to identify major gaps in knowledge and to define confidence intervals for the results obtained (Erisman, 1992; 1993a).

8. Gaps in knowledge

The present state of knowledge on SO_2 dry deposition mechanisms has not been translated into parametrizations for transport models so far. It is doubtful whether the present state of knowledge and the availability of data allows such a detailed parametrization. No full coverage on R_c values for all types of surfaces and their time varia-

tions exist. There is a great need for mechanistic studies to provide the information for these parametrizations. These studies should focus on the influence of water films, their composition and evaporation, the influence of neutralising processes (co-deposition, canopy leaching) and the influence of type of vegetation on SO_2 dry deposition. The difficulty in controlling or measuring the wide range of variables which all influence water film chemistry, and reduction of these complex process into simple parametrizations will be one of the most challenging tasks in future SO_2 dry deposition research. The success of the model concept to estimate SO_2 dry deposition on a local scale in Europe, as presented in this paper strongly depends on the availability of data. Land use data covering the whole of Europe are available on a 0.5×0.5 latitude-longitude grid (Olson et al., 1985). Data on a smaller scale are available only for some countries. The resolution of the land use database has to be improved. The concept of the roughness length is only valid for horizontal homogeneous surfaces. Most of the landscapes in Europe, however, represent so called complex terrain, i.e., many roughness transition zones such as forest edges; hills and mountains. It has to be investigated whether the roughness length concept can be applied in these areas. Orographic effects can induce large errors in the modelled concentration/deposition since these effects are not yet accounted for in models.

If co-deposition is the main process in SO_2 dry deposition during surface wetness, measuring or modelling SO_2 deposition alone will probably not be enough. Each component depositing into a water film and the origin of the water film will determine its chemistry. Therefore, dry deposition of HNO_3, HNO_2, HCl, NH_3, small and coarse particles, and oxidising species should be modelled on the same time scales. At this moment there is no approach available for such an exercise. Furthermore there is a serious lack of information to develop it.

There is a serious lack of data needed for estimating soil moisture and surface wetness and its origin (rain, fog, cloud, dew, guttation). Emission should become available on a smaller scale than 50×50 km for the whole of Europe. Information on emission variations in time is needed. The model concept proposed in this paper might be used for other components. However, it has to be investigated whether it is valid for components showing horizontal variations in concentrations due to the release by ground-level sources (NH_3, NO_x) and for components which can both be emitted and deposited at the earth surface.

9. Acknowledgements

Bruce Hicks (NOAA ARL, USA), David Fowler and Mark Sutton (ITE, UK), Frank de Leeuw, Hans van Jaarsveld and Addo van Pul (RIVM) are kindly acknowledged for their review of this paper. Jürgen Kesselmeier is acknowledged for providing the SO_2 emission data.

REFERENCES

Andreae, M. O. 1991. Biomass burning: its history, use and distribution and its impact on environmental quality and global climate. In: *Global biomass burning: atmospheric, climatic and biospheric implications* (ed. Levine, J. S.), MIT Press, Boston, pp. 3–21.

Andreae, M. O. and Andreae, T. W. 1988. The cycle of biogenic sulfur compounds over the Amazon basin. 1. Dry season. *J. Geophys. Res.* **93**, 1487–1497.

Andreae, M. O. and Jaeschke, W. A. 1992. Exchange of sulphur between biosphere and atmosphere over temperate and tropical regions. In: *Sulphur cycling on the continents* (eds. Howarth, R. W., Stewart, J. W. B. and Ivanow, M. V.), John Wiley and Sons Ltd., pp. 27–61.

Andreae, M. O. and Raemdonck, H. 1983. Dimethylsulfide in the surface ocean and the marine atmosphere. *Science* **221**, 744–777.

Baldocchi, D. D., Hicks, B. B. and Camara, P. 1987. A canopy stomatal resistance model for gaseous deposition to vegetated surfaces. *Atmospheric Environment* **21**, 91–101.

Baldocchi, D. D., Hicks, B. B. and Meyers, T. P. 1988. Measuring biosphere-atmosphere exchanges of biologically related gases with micrometeorological methods. *Ecology* **69**, 1331–1340.

Barrie, L. A. and Warmsley, J. L. 1978. A study of sulphur dioxide deposition velocities to snow in northern Canada. *Atmospheric Environment* **12**, 2321–2322.

Bates, T. S., Cline, J. D., Gammon, R. H. and Kelly-Hansen, S. R. 1987. Regional and seasonal variations in the flux of oceanic dimethylsulfide to the atmosphere. *J. Geophys. Res.* **92**, 2930–2938.

Berresheim, H., Andreae, M. O., Ayers, A. P. and Gillet,

R. W. 1989. Distribution of biogenic sulfur compounds in the remote southern hemisphere. In: *Biogenic sulfur in the environment* (Saltzmann, E. S. and Cooper, W. J., eds.), ACS Symposium Series, ISBN 0-8412-1612-6, pp. 352–366.

Brimblecombe, P. 1978. "Dew" as a sink for sulphur dioxide. *Tellus* **30**, 151–157.

Businger, J. A. 1986. Evaluation of the accuracy with which dry deposition can be measured with current micrometeorological techniques. *J. Climate Appl. Meteor.* **25**, 1100–1124.

Caddle, S. H., Dasch, J. M. and Mulawa, P. A. 1985. Atmospheric concentrations and the deposition velocity to snow of nitric acid, sulphur dioxide, and various particulate species. *Atmospheric Environment* **19**, 1819–1827.

Chamberlain, A. C. 1980. Dry deposition of sulphur dioxide. In: *Atmospheric sulphur deposition* (eds. Shriner, D. S., Richmond, C. R. and Lindberg, S. E.), Ann Arbor Science, Michigan, USA, 185–197.

Chameides, W. L. 1987. Acid dew and the role of chemistry in the dry deposition of reactive gases to wetted surfaces. *J. Geophys. Res.* **92**, 11895–11908.

Collatz, G. J., Ball, J. T., Grives, C. and Berry, J. A. 1991. Regulation of stomatal conductance and transpiration: a physiological model of canopy processes. *Agr. For. Meteor.* **54**, 107–136.

Cullis, C. F. and Hirschler, M. M. 1980. Atmospheric sulfur: Natural and man-made sources. *Atmospheric Environment* **14**, 1263–1278.

Davidson, C. I. and Wu, Y. L. 1989. Dry deposition of particles and vapors. In: *Acidic precipitation*, vol. 3, eds. Lindberg, S. E., Page, A. L. and Norton, S. A., Springer-Verlag, Berlin, 103–209.

Davies, T. D. and Mitchell, J. R. 1983. Dry deposition of sulphur dioxide onto grass in rural Eastern England with some comparison with other forms of sulphur deposition. In: *Precipitation scavenging, dry deposition and resuspension.* Pruppacher et al. (eds.). Elsevier Science Publishing Co., Inc., 795–806.

Davies, T. D. and Wright, R. G. 1985. Sulphur dioxide deposition velocity by a concentration gradient measurement system. *J. Geophys. Res.* **90**, 2091–2095.

Dop, H. van 1983. Terrain classification and derived meteorological parameters for inter-regional transport models. *Atmospheric Environment* **17**, 1099–1105.

Draaijers, G. P. J. and Erisman, J. W. 1993. Atmospheric sulphur deposition onto forest stands: throughfall estimates compared to estimates from inference. *Atmospheric Environment* **27A**, 43–55.

Duyzer, J. H. and Bosveld, F. C. 1988. Measurements of dry deposition fluxes of O_3, NO_x, SO_2 and particles over grass/heathland vegetation and the influence of surface inhomogeneity. *Report no. R 88/111.* TNO, Delft, the Netherlands.

Erisman, J. W. 1992. *Atmospheric deposition of acidifying compounds in The Netherlands.* Ph.D. Thesis, University of Utrecht, Utrecht, the Netherlands.

Erisman, J. W. 1993a. Acid deposition onto nature areas in the Netherlands; Part I. Methods and results. *Water, Soil and Air Pollut.* **71**, 51–80.

Erisman, J. W. 1993b. Acid deposition onto nature areas in the Netherlands; Part II. Throughfall measurements compared to deposition estimates. *Water, Soil and Air Pollut.* **71**, 81–99.

Erisman, J. W., Leeuw, F. A. A. M. de and Aalst, R. M. van 1989. Deposition of the most acidifying compounds in the Netherlands during 1980–1986. *Atmospheric Environment* **23**, 1051–1062.

Erisman, J. W. and Wyers, G. P. 1993. On the interaction between deposition of SO_2 and NH_3. *Atmospheric Environment* **27A**, 1937–1949.

Erisman, J. W., Versluis, A. H., Verplanke, T. A. J. W., de Haan, D., Anink, D., van Elzakker, B. G., Mennen, M. G. and Aalst, R. M. van 1993a. Monitoring the dry deposition of SO_2 in the Netherlands. *Atmospheric Environment* **27**, 1153–1161.

Erisman, J. W., Beier, C., Draaijers, G. and Lindberg, S. 1994. Review of deposition monitoring methods. *Tellus* **46**, 79–93.

Erisman, J. W., Elzakker, B. G. van, Mennen, M. G., Hogenkamp, J., Zwart, E., Beld, L. van den, Römer, F. G., Bobbink, R., Raessen, M., Duyzer, J. H. and Verhage, H. L. M. 1993c. The Elspeetsche Veld experiment on surface exchange of trace gases: summary of results. *Atmospheric Environment*, in press.

Erisman, J. W., Mennen, M., Hogenkamp, J., Kemkers, E., Goedhart, D., Pul, A. van and Boermans, J. 1993d. Dry deposition over the Speulder forest. *Proceedings of the CEC/BIATEX workshop*, Aveiro, Portugal, 4–7 May 1993.

Finlayson-Pitts, B. J. and Pitts, J. M. 1986. *Atmospheric chemistry.* John Wiley and Sons, New York.

Fowler, D. 1978. Dry deposition of SO_2 on agricultural crops. *Atmospheric Environment* **12**, 369–373.

Fowler, D. 1981. Turbulent transfer of sulphur dioxide to cereals: a case study. In: *Plants and flux*, pp. 139–146.

Fowler, D. 1985. Dry deposition of SO_2 onto plant canopies. In: *Sulphur dioxide and vegetation* (eds. Winner, W. E., Mooney, H. A. and Goldstein, R. A.). Stanford University Press, California, 75–95.

Fowler, D. and Unsworth, M. H. 1979. Turbulent transfer of sulphur dioxide to a wheat crop. *Quart. J. R. Met. Soc.* **105**, 767–783.

Fowler, D. and Cape, J. N. 1984. The contamination of rain samples by dry deposition on rain collectors. *Atmospheric Environment* **18**, 183–189.

Fowler, D. and Duyzer, J. H. 1990. Micrometeorological techniques for the measurement of trace gas exchange. In: *Exchange of trace gases between terrestrial ecosystems and the atmosphere*, eds. Andreae, M. O. and Schimel, D. S., John Wiley and Sons, 189–207.

Fowler, D., Duyzer, J. H. and Baldocchi, D. D. 1991. Inputs of trace gases, particles and cloud droplets to terrestrial surfaces. In: *Proc. of the R. Soc. of Edinburgh* **97B**, 35–59.

Galbally, I. 1979. Sulphur uptake from the atmosphere by forest and farmland. *Nature* **280**, 49–50.

Garland, J. A. 1977. The dry deposition of sulphur dioxide to land and water surfaces. *Proc. R. Soc. Lond. A* **354**, 245–268.

Garland, J. A. 1978. Dry and wet removal of sulfur from the atmosphere. *Atmospheric Environment* **12**, 349.

Garland, J. A. and Branson, J. R. 1977. The deposition of sulphur dioxide to pine forest assessed by a radioactive tracer method. *Tellus* **29**, 445–454.

Garsed, S. G. 1985. SO_2 uptake and transport. In: *Sulphur dioxide and vegetation* (eds. Winner, W. E., Mooney, H. A. and Goldstein, R. A.). Stanford University Press, California, 75–95.

Granat, L. and Johansson, C. 1983. Dry deposition of SO_2 and NO_x in winter. *Atmospheric Environment* **17**, 191–192.

Hicks, B. B., Wesely, M. L., Durham, J. L. and Brown, M. A. 1982. Some direct measurements of atmospheric sulphur fluxes over a pine plantation. *Atmospheric Environment* **16**, 2899–2903.

Hicks, B. B., Wesely, M. L., Coulter, R. L., Hart, R. L., Durham, J. L., Speer, R. E. and Stedman, D. H. 1983. An experimental study of sulphur deposition to grassland. In: *Precipitation scavenging*, dry deposition and resuspension (eds. Pruppacher et al.). Elsevier Science Publishing, Amsterdam, pp. 933–942.

Hicks, B. B., Baldocchi, D. D., Meyers, T. P., Hosker, Jr. R. P. and Matt, D. R. 1987. A preliminary multiple resistance routine for deriving dry deposition velocities from measured quantities. *Water, Air and Soil Pollut.* **36**, 311–330.

Hicks, B. B., Wesely, M. L., Coulter, R. L., Hart, R. L., Durham, J. L., Speer, R. E. and Stedman, D. H. 1983. An experimental study of sulphur and NO_x fluxes over grassland. *Boundary-Layer Meteor.* **34**, 103–121.

Hicks, B. B., Matt, D. R. and McMillen, R. T. 1989. A micrometeorological investigation of surface exchange of O_3, SO_2 and NO_2: a case study. *Boundary-Layer Meteor.* **47**, 321–336.

Hicks, B. B., Draxler, R. R., Albritton, D. L., Fehsenfeld, F. C., Hales, J. M., Meyers, T. P., Vong, R. L., Dodge, M., Schwartz, S. E., Tanner, R. L., Davidson, C. I., Lindberg, S. E. and Wesely, M. L. 1989. *Atmospheric processes research and processes model development.* State of Science/Technology report no. 2. National Acid Precipitation Assessment Program, NAPAP office, Washington, USA.

Hove, L. W. A. van 1989. *The mechanism of NH_3 and SO_2 uptake by leaves and its physiological effects.* Thesis, University of Wageningen, the Netherlands.

Jaarsveld, H. J. A. and Onderdelinden, D. 1990. *TREND: An analytical long term deposition model for multi-scale applications.* Report no. 228603009, National Institute of Public Health and Environmental Protection, Bilthoven, the Netherlands.

Jarvis, P. G. 1976. The interpretation of the variations in leaf water potential and stomatal conductance found in canopies in the field. *Phyl. Trans. R. Soc. Lond.* **B273**, 593–610.

Iversen, T., Halvorsen, N., Mylona, S. and Sandnes, H. 1991. *Calculated budgets for airborne acidifying compounds in Europe, 1985, 1987, 1988 and 1990.* Meteorological synthesizing Centre-West, the Norwegian Meteorological Institute, Oslo, Norway.

Lorenz, R. and Murphy, C. H. Jr. 1985. Sulphur dioxide, carbon dioxide, and water vapour flux measurements utilizing a microprocessor-controlled data acquisition system in a pine plantation. In: *The forest-atmosphere interaction* (eds. Hutchison, B. A. and Hicks, B. B.). Kluwer, Dordrecht, The Netherlands, 133–148.

Lövblad, G. and Erisman, J. W. 1992. Deposition of nitrogen in Europe. In: *Critical loads for nitrogen* (eds. Grennfelt, P. and Thörnelöf, E.). Lökeberg, 6–10 April 1992, Sweden, Report no. Nord 1992:41, Nordic Council of Ministers, Copenhagen, Denmark, 239–286.

Matt, D. R., McMillen, R. T., Womack, J. D. and Hicks, B. B. 1987. A comparison of estimated and measured SO_2 deposition velocities. *Water, Air and Soil Pollut.* **36**, 331–347.

McMillen, R. T., Matt, D. R., Hicks, B. B. and Womack, J. D. 1987. Dry deposition measurements of Sulphur dioxide to a Spruce-Fir forest in the Black forest: A data report. NOAA Technical Memorandum ERL ARL-152.

Meyers, T. P. 1987. The sensitivity of modelled SO_2 fluxes and profiles to stomatal and boundary layer resistance. *Water, Air and Soil Pollut.* **35**, 261–278.

Meyers, T. P. and Baldocchi, D. D. 1988. A comparison of models for deriving dry deposition fluxes of O_3 and SO_2 to a forest canopy. *Tellus* **40B**, 270–284.

Milne, J. W., Roberts, D. B. and Williams, D. J. 1979. *Atmospheric Environment* **13**, 373–379.

Murphy, C. E., Sinclair, T. R. and Knoerr, K. R. 1977. An assessment of the use of forests as sinks for the removal of atmospheric sulphur dioxide. *J. Environ. Qual.* **6**, 388–396.

Murphy, C. E. and Sigmon, J. T. 1989. Dry deposition of sulphur and nitrogen oxide gases to forest vegetation. In: *Acidic precipitation*, vol. 3, eds. Lindberg, S. E., Page, A. L. and Norton, S. A., Springer-Verlag, Berlin, 217–238.

Neumann, H. H. and Hartog, C. D. 1985. Eddy correlation measurements of atmospheric fluxes of ozone, sulphur, and particles during the campaign intercomparison study. *J. Geophys. Res.* **90**, 2097–2110.

Nicholson, K. W. and Davies, T. D. 1988. Letter to the editors: The dry deposition of sulphur dioxide at a rural site. *Atmospheric Environment* **22**, 2885–2889.

O'Dell, R. A., Taheri, M. and Kabel, R. L. 1973. A model for uptake of pollutants by vegetation. *JAPCA* **27**, 1104–1109.

Payrissat, M. and Beilke, S. 1975. Laboratory measurements of the uptake of sulphur dioxide by different European soils. *Atmospheric Environment* **9**, 211–217.

Platt, U. 1978. Dry deposition of SO_2. *Atmospheric Environment* **12**, 363–367.

Petit, C., Ledoux, M. and Trinite, M. 1977. Transfer

resistance to SO_2 capture by some plant surfaces, water and leaves. *Atmospheric Environment* **11**, 1123–1126.

Pul, W. A. J. van, Erisman, J. W., Jaarsveld, J. A. van and Leeuw, F. A. A. M. de. High resolution assessment of acid deposition fluxes. In: *Acidification research: evaluation and policy application*, ed. T. Schneider. Studies in Environmental Science, Elsevier, Amsterdam, 277–284.

Ruigrok, W. and Davidson, C. I. 1992. Particle deposition. In: *Models and methods for the quantification of atmospheric input to ecosystems* (eds. Lövblad, G., Erisman, J. W. and Fowler, D.). Nordiske Seminar- of Arbejdsrapporter 1993:573, The Nordic Council of Ministers, Copenhagen, 145–162.

Schwela, D. 1977. Die trockene Deposition gasförmiger Luftverunreinigungen. Schriftenreihe de Landesanstalt für Immissionsschutz. *Heft* **42**, 46–85.

Seinfeld, J. H. 1986. *Atmospheric chemistry and physics of air pollution.* John Wiley and Sons, New York.

Taylor, G. E., McLaughin, S. B., Shriner, D. S. and Selvidge, W. J. 1983. The flux of sulphur containing gases to vegetation. *Atmospheric Environment* **17**, 789–796.

Taylor, G. E. and Tingley, D. T. 1983. Sulphur dioxide flux into leaves of *Geranium carolinianum* L. *Plant Physiology* **72**, 237, 244.

Thom, A. S. 1975. Momentum, mass and heat exchange of plant communities. In: *Vegetation and atmosphere*, pp. 58–109 (ed. Monteith, J. L.). London, Academic Press.

Thomas, R., Arkel, W. G. van, Baars, H. P., Ierland, E. C., Boer, K. F. de, Buijsman, E., Hutten, T. J. H. M. and Swart, R. J. 1988. *Emissions of SO_2, VOC and NH_3 in the Netherlands and in Europe in the period 1950–2030.* Report no. 758472002. National Institute of Public Health and Environmental Protection, Bilthoven, the Netherlands.

UK Review Group on Acid Rain 1990. *Acid deposition in the United Kingdom.* Warren Spring Laboratory, Stevenage, UK.

Valdez, M. P., Bales, R. C., Stanley, D. A. and Dawson, G. A. 1987. Gaseous deposition to snow 1. Experimental study of SO_2 and NO_2 deposition. *J. Geophys. Res.* **92**, 9779–9787.

Vermetten, A. W. H., Hofschreuder, P., Duyzer, J. H., Diederen, H. S. M. A., Bosveld, F. C. and Bouten, W. 1992. Dry deposition of SO_2 onto a stand of Douglas fir: the influence of canopy wetness. In: *Proc. 5th IPSASEP conference*, Richland, 15–19 June 1991 USA, eds. Schwartz, S. E. and Slinn, W. G. N. Hemisphere Publishing Corporation. Washington, 1403–1414.

Voldner, E. C., Barrie, L. A. and Sirois, A. 1986. A literature review of dry deposition of oxides of sulfur and nitrogen with emphasis on long-range transport modelling in North America. *Atmospheric Environment* **20**, 2101–2123.

Welphdale, D. M. and Shaw, R. W. 1974. Sulphur dioxide removal by turbulent transfer over grass, snow, and water surfaces. *Tellus* **26**, 196–204.

Welphdale, D. M. 1987. Atmospheric deposition of acidic pollutants. In: *Int. Symp. on Acidification and Water Pathways*, Bolkesjø, Norway, 4–8 May, 1987.

Wesely, M. L. and Hicks, B. B. 1977. Some factors that affect the deposition rates of Sulfur Dioxide and similar gases on vegetation. *J. Air. Poll. Contr. Ass.* **27**, 1110–1116.

Wesely, M. L. 1989. Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. *Atmospheric Environment* **23**, 1293–1304.

Wesely, M. L., Sisteron, D. L. and Jastrow, J. D. 1990. Observations of the chemical properties of dew on vegetation that affect the dry deposition of SO_2. *J. Geophys. Res.* **95**, 7501–7514.